Antireflection of an absorbing substrate by an absorbing thin film at normal incidence

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An absorbing substrate of complex refractive index \( n_2 - jk_2 \) at wavelength \( \lambda \) can be coated by an absorbing thin film of complex refractive index \( n_1 - jk_1 \) and thickness \( d \) to achieve zero reflection at normal incidence. For given \( n_2, k_2 \), multiple solutions \((n_1, k_1, d/\lambda)\) are found that correspond to infinitely many distinct antireflection layers. This is demonstrated for a Si substrate at two wavelengths (6328 and 4420 Å). The response of these absorbing antireflection layers to changes of the angle of incidence from 0 to 45° and to changes of thickness of ±10% is also determined and compared to the limiting case of a nonabsorbing antireflection layer.

I. Introduction

An absorbing substrate can be coated by a transparent thin film for zero reflection of normally incident monochromatic light, as first noted by Hass et al.\(^1\) and subsequently by Park.\(^2\) In this case, if \( N_2 = n_2 - jk_2 \) is the complex refractive index of the substrate and \( N_1 = n_1 \) is the real refractive index of the transparent film at wavelength \( \lambda \), it can be shown\(^1,2\)

\[
n_{1t} = \left( n_2 + \frac{k_2^2}{n_2 - 1} \right)^{1/2},
\]

where incidence from air \((N_0 = 1)\) is assumed. The associated required film thickness is given by

\[
d = (\xi + m)D_s,
\]

where

\[
\xi = (1/2\pi) \arctan[2n_{1t}k_2/(n_{1t} - n_2 - k_2)],
\]

\[
D_s = \lambda/2n_{1t}
\]

is the film thickness period, and \( m \) is an integer.

The suggestion of using absorbing thin films as antireflection coatings on metals was also due to Hass et al.\(^1\) The need to do so arises because it is often desirable to coat a metal substrate with a derivative oxide film that is often nonstoichiometric, hence absorbing. Examples of this are titanium oxide films on Ti,\(^3\) iron oxide films on Fe (or steel), and scandium oxide films on Sc.\(^4\) The primary application of these coatings is for solar energy collection.\(^5\)

This paper presents an analytical and numerical study of normal-incidence antireflection of an absorbing substrate by an absorbing thin film. For a given substrate of complex refractive index \( N_2 = n_2 - jk_2 \), multiple solutions are determined for the film properties \((n_1, k_1, d/\lambda)\), where \( n_1 - jk_1 \) is the film complex refractive index and \( d/\lambda \) is its normalized thickness. (The presence of these multiple solutions was apparently missed in previous accounts of this problem.) Furthermore, we consider the response of these antireflection coatings to changes of angle of incidence from 0 to 45° and to changes of thickness (or equivalently of wavelength, neglecting material dispersion) of ±10%. For illustration, a concrete example is considered of absorbing antireflection layers on a Si substrate at wavelengths \( \lambda = 6328 \) Å (of the He–Ne laser) and 4420 Å (of the He–Cd laser). The choice of Si is clearly justified by its common use in photodetectors and solar cells.

II. Antireflection Condition and Multiple Solutions

The reflection coefficient for normally incident monochromatic light of a film with plane-parallel boundaries on a substrate is given by

\[
R = (r_{01} + r_{12}X)/(1 + r_{01}r_{12}X),
\]

independent of polarization. \( r_{01} \) and \( r_{12} \) are the ambient–film and film–substrate interface Fresnel complex reflection coefficients, respectively, and are given by

\[
r_{01} = (1 - N_1)/(1 + N_1), \quad r_{12} = (N_1 - N_2)/(N_1 + N_2),
\]

where \( N_1 \) and \( N_2 \) are the complex refractive indices of the (homogeneous, linear, optically isotropic, and nonmagnetic) media of the film and substrate, respective-
ly, and incidence from air \((N_0 = 1)\) is assumed. \(X\) is a complex exponential function of film thickness \(d\) given by

\[
X = \exp(-4j\pi N_1 d/\lambda) \cdot \exp(j2\pi m).
\]  

(7)

The second term in Eq. (7), where \(m\) is an integer, is equal to 1, and its addition is the key to finding all possible solutions.

Antireflection occurs when

\[
R = 0
\]

or

\[
r_{01} + r_{12} X = 0
\]

from Eq. (5). From Eq. (9) one gets

\[
X = -r_{01}/r_{12}
\]

(10)

Substitution of Eqs. (6) and (7) into Eq. (10), taking the natural logarithm of both sides, and solving the resulting equation for \(d/\lambda\), we obtain

\[
d/\lambda = (\pi/\lambda N_1) \ln[1 - \exp(-j\pi M)] - \ln(1 + N_1)
\]

\[\quad - \ln(N_1 - N_2) + \ln(N_1 + N_2),\]

(11)

where \(M = 2m \pm 1\) is an odd integer.

For a given \(N_2 = n_2 - jk_2\), Eq. (11), whose right-hand side is a complex function of \(N_1 = n_1 - jk_1\), can be rewritten as

\[
d/\lambda = f_M(n_1,k_1),
\]

(12)

where the subscript \(M\) is the same integer that appears in Eq. (11). Next Eq. (12) is broken into its real and imaginary parts as follows:

\[
\text{Im}[f_M(n_1,k_1)] = 0,
\]

(13)

\[
\text{Re}[f_M(n_1,k_1)] = d/\lambda.
\]

(14)

Equation (13) represents the necessary constraint on the optical constants \(n_1,k_1\) of the antireflection layer. This can be represented by a family of curves in the \(n_1k_1\) plane corresponding to different values of the integer \(M\). As will be demonstrated in the following section, solutions of Eq. (13) exist for positive odd integers \(M\) and only for \(n_1 \leq n_{1tt}\), where \(n_1 = n_{1tt}\) is given by Eq. (1) and is associated with \(k_1 = 0\), i.e., the limiting case of a transparent film.

For a given set \(M,n_1,k_1\) that satisfies Eq. (13), the associated normalized thickness \(d/\lambda\) is obtained subsequently by direct calculation from Eq. (14).

### III. Numerical Examples

Consider a silicon substrate with \(N_2 = 3.884 - j0.02\) at \(\lambda = 6328\ \text{Å}\). Figure 1 shows a family of solution curves of Eq. (13) for \(M = 1, 3, 5, 7,\) and \(9\) in the \(n_1k_1\) plane. All curves meet at a common point on the \(n_1\) axis \((k_1 = 0)\), where \(n_1 = n_{1tt} = 1.970821\), as is obtained by substituting \(n_2 = 3.884, k_2 = 0.02\) in Eq. (1). This is the refractive index of the unique transparent antireflection layer and fortuitously corresponds to that of stoichiometric \(\text{Si}_3\text{N}_4\). No solutions exist when \(n_1 > n_{1tt}\), and for a given \(M\) (e.g., \(M = 1\)), \(k_1\) increases as \(n_1\) decreases below the limit \(n_{1tt}\) over the range of \(n_1\) values \((1.25 \leq n_1 \leq 1.970821)\) shown in Fig. 1.

The normalized thickness \(d/\lambda\) of the absorbing antireflection layer on Si is plotted vs \(n_1\) in Fig. 2 corresponding to each and every solution curve in Fig. 1 as identified by the integer \(M\). It is evident that the thinnest antireflection layers are associated with the largest values of the film extinction coefficient \(k_1\) (case of \(M = 1\)). As \(M\) increases, \(k_1\) decreases, and \(d/\lambda\) increases.

To cite some specific numerical results, Table I lists \(k_1\) and \(d/\lambda\) when \(n_1 = 1.9\) for \(M = 1, 3, 5, 7,\) and \(9\). The film optical constants \(n_1,k_1\) given in this table appear

![Figure 1](image1.png)

***Fig. 1.*** Constraint on the optical constants \(n_1,k_1\) of an absorbing layer on a Si substrate \((n_2 = 3.884, k_2 = 0.02)\) that produce antireflection at normal incidence for light of wavelength \(\lambda = 6328\ \text{Å}\). Multiple-solution branches are obtained from Eq. (13) for order numbers \(M = 1, 3, 5, 7,\) and \(9\).

![Figure 2](image2.png)

***Fig. 2.*** Variation of the normalized film thickness \(d/\lambda\) of the absorbing antireflection layer on Si as a function of film refractive index \(n_1\) along each solution branch in Fig. 1, as obtained from Eq. (14).

### Table I. Extinction Coefficient \(k_1\) and Normalized Thickness \(d/\lambda\) of Several Absorbing Antireflection Layers with \(n_1 = 1.9\) on a Si Substrate at \(\lambda = 6328\ \text{Å}\)

<table>
<thead>
<tr>
<th>(M)</th>
<th>1</th>
<th>3</th>
<th>5</th>
<th>7</th>
<th>9</th>
</tr>
</thead>
<tbody>
<tr>
<td>(k_1)</td>
<td>0.058033</td>
<td>0.020080</td>
<td>0.012088</td>
<td>0.08642</td>
<td>0.006724</td>
</tr>
<tr>
<td>(d/\lambda)</td>
<td>0.134806</td>
<td>0.395673</td>
<td>0.658348</td>
<td>0.921298</td>
<td>1.184340</td>
</tr>
</tbody>
</table>

*The complex refractive index of Si at this wavelength is assumed to be 3.884–j0.02. \(M\) is the order number that appears in Eq. (11).*
Fig. 3. Same as in Fig. 1 for \( \lambda = 4420 \) Å. At this wavelength \( n_2 = 4.775 \) and \( k_2 = 0.161 \) for the Si substrate.

Fig. 4. Normalized film thickness \( d/\lambda \) vs film refractive index \( n_1 \) for each solution curve in Fig. 3.

to be realizable by controlling the formation (deposition) conditions and stoichiometry of the silicon nitride film on Si.

If the wavelength is changed to 4420 Å, the absorption of Si significantly increases, and its complex refractive index becomes \( N_2 = 4.775-j0.161 \). The solutions for antireflection layers correspondingly change and become those given in Figs. 3 and 4. The optical constants of Si that we use are obtained by interpolation from the data presented by Aspnes and Studna. The curve for the limiting case of a transparent antireflection film (\( n_1 = 1.970821, k_1 = 0 \)) is added in Fig. 5 but is indistinguishable from that of \( n_1 = 1.9, k_1 = 0.058033 \). \( R_u = 0 \) at \( \phi = 0 \) for all solutions, but it increases to unacceptably high levels at oblique incidence for the thicker antireflection layers (\( M = 5, 7, \) and 9). At \( \phi = 45^\circ \), \( R_u = 1.87, 6.35, 13.23, 20.22, \) and 25.95% when \( M = 1, 3, 5, 7, \) and 9, respectively. The thinnest absorbing film has the best angular response and is the one to use for light incident within a wide (45° semiapex angle) cone. It is significant to note again that such a film is virtually as good as the perfectly dielectric antireflection film (for which \( R_u = 1.83\% \) at \( \phi = 45^\circ \)).

IV. Angular and Thickness Sensitivity

For a given antireflection layer on Si we calculated the reflectance

\[
R_u = \frac{(R_p + R_s)}{2},
\]

(15)

for incident unpolarized light as a function of (1) the angle of incidence \( \phi \) from 0 to 45° and (2) the film thickness ratio \( d/d_0 \) between 0.9 and 1.1, where \( d_0 \) is the antireflection thickness.

Figure 5 shows \( R_u \) vs \( \phi \) for the five solutions at \( n_1 = 1.9 \) that correspond to \( M = 1, 3, 5, 7, \) and 9 (Table I).

The curve for the limiting case of a transparent antireflection film (\( n_1 = 1.970821, k_1 = 0 \)) is added in Fig. 5 but is indistinguishable from that of \( n_1 = 1.9, k_1 = 0.058033 \). \( R_u = 0 \) at \( \phi = 0 \) for all solutions, but it increases to unacceptably high levels at oblique incidence for the thicker antireflection layers (\( M = 5, 7, \) and 9). At \( \phi = 45^\circ \), \( R_u = 1.87, 6.35, 13.23, 20.22, \) and 25.95% when \( M = 1, 3, 5, 7, \) and 9, respectively. The thinnest absorbing film has the best angular response and is the one to use for light incident within a wide (45° semiapex angle) cone. It is significant to note again that such a film is virtually as good as the perfectly dielectric antireflection film (for which \( R_u = 1.83\% \) at \( \phi = 45^\circ \)).

Figure 6 shows \( R_u \) vs \( d/d_0 \) for the same five solutions of Table I and for the reference case of the transparent antireflection film on Si. The thinnest absorbing antireflection layer with \( n_1 = 1.9, k_1 = 0.058033 \) has virtually the same response to thickness changes as the transparent antireflection layer with \( n_1 = 1.970821 \) and \( k_1 = 0 \) (the two coincident bottom curves). The response to thickness variation of the higher-order (\( M \geq 3 \)) solutions with thicker less-absorbing films is evidently unacceptable.

It is also worthwhile to consider the angular and thickness sensitivity of the smallest-thickness films (\( M = 1 \)) as \( n_1 \) is decreased (and \( k_1 \) is increased). For reference, Table II summarizes data for four such solutions that correspond to \( n_1 = 1.5, 1.7, 1.9, \) and 1.970821.
Table II. Refractive Index \( n_1 \), Extinction Coefficient \( k_1 \), and Normalized Thickness \( d/\lambda \) of Four Antireflection Layers of the Lowest Possible Thickness (Order Number \( M = 1 \)) on a Si Substrate (\( n_2 = 3.884, k_2 = 0.02 \)) at \( \lambda = 6328 \) Å

<table>
<thead>
<tr>
<th>( n_1 )</th>
<th>( k_1 )</th>
<th>( d/\lambda )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.970821</td>
<td>0</td>
<td>0.126851</td>
</tr>
<tr>
<td>1.9</td>
<td>0.058033</td>
<td>0.134806</td>
</tr>
<tr>
<td>1.7</td>
<td>0.187869</td>
<td>0.161414</td>
</tr>
<tr>
<td>1.5</td>
<td>0.273651</td>
<td>0.195963</td>
</tr>
</tbody>
</table>

Figure 7 illustrates \( R_u \) vs \( \phi \) for the four antireflection layers of Table II. All curves are closely spaced, and the angular response of the nonabsorbing film is only slightly better than that of the absorbing ones.

Finally, Fig. 8 depicts \( R_u \) vs \( d/d_0 \) for the same four antireflection layers of Table II. The performance is good in all cases and steadily improves as \( n_1 \) is decreased and \( k_1 \) is increased. Thus, from the point of view of thickness (wavelength) sensitivity, absorbing antireflection films offer a modest advantage. It is also interesting to note the asymmetry of the \( R_u \) vs \( d/d_0 \) curve (around \( d/d_0 = 1 \)) in Fig. 8 and its change as \( n_1 \) is decreased.

V. Summary

The problem of antireflection coating of an absorbing substrate using an absorbing thin film has been covered here more completely than before. For a given substrate with known optical constants \( n_2, k_2 \) at a given wavelength, (heretofore undiscovered) multiple solutions \( (n_1, k_1, d/\lambda) \) are found for an absorbing thin-film coating that produces zero reflection at normal incidence. This should facilitate achieving antireflection of absorbing substrates. These multiple solutions are displayed as a family of curves in the \( n_1, k_1 \) plane for a Si substrate at two wavelengths to provide specific numerical examples of the analytically derived antireflection conditions. Furthermore, we have considered the angular and thickness response of several absorbing antireflection layers and compared the results with the limiting case of a transparent film.

R. M. A. Azzam was with the Marseilles group as an invited professor and senior Fulbright research scholar when this work was completed.

References

4. Optical constants of nonstoichiometric scandium oxide films and of Sc obtained in one of our laboratories (Marseille) indicate the utility of this material system as a good solar absorber.
5. In the VUV spectral region transparent thin-film coating materials are lacking, and antireflection by absorbing films is again important, e.g., for efficient photodetection.